

Response Under 37 C.F.R. §1.116

Expedited Procedure

Examining Group 1792

Application No. 10/564,015

Paper Dated: February 10, 2009

In Reply to USPTO Correspondence of December 10, 2008

Attorney Docket No. 0149-053929

REMARKS

Claim 1 has been amended to recite the feature that the hydrolysable metal compound vapor and the water vapor are reactant with one another. Claim 1 has also been amended to incorporate the features of dependent claim 2 to clarify that the vapor deposition step is carried out by injection of jetted streams of the hydrolysable metal compound vapor and water vapor and that the mixing is performed by injecting the hydrolysable metal compound vapor and the water vapor in such a manner that the resulting two jetted vapor streams meet each other *before* they reach the substrate. The dependency of claims 3-4 has been amended since claim 2 has been canceled. These claims now depend from claim 1.

Support for these amendments is provided throughout the specification and claims as originally filed and specifically at page 6, lines 27-29, page 11, lines 2-8 and Figure 1.

No new matter has been added.

It is respectfully requested that this Amendment be entered as it does not introduce new issues that require further search and/or consideration. The issue of the hydrolysable metal compound vapor and water vapor being reactant with one another and the problems associated with this reactivity is the subject matter of the present application. Furthermore, the reactivity of these components is discussed on page 8 of the Response filed September 15, 2008. Dependent claim 2, which previously depended from independent claim 1, has already been examined and the subject matter recited therein has been fully incorporated into independent claim 1.

The Examiner's reconsideration is respectfully requested in light of the amendments made herein, taken with the following remarks.

RESPONSE TO REJECTIONS

The present invention is directed to a process for forming a metal oxide film comprising a vapor deposition step in which a vapor of a hydrolysable metal compound and water vapor are brought into contact with a substrate to form a film of a metal oxide precursor on the surface of the substrate and a calcination step in which the substrate is then heated in an oxidizing atmosphere to convert the precursor into a metal oxide. During the vapor deposition

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step, the hydrolysable metal compound vapor and the water vapor are mixed and, due to the high reactivity of the metal compound vapor and the water vapor, these mixed vapors are brought into contact with the substrate within three seconds after mixing. By bringing the hydrolysable metal compound vapor and water vapor into contact with a substrate within three seconds of mixing, the occurrence of a hydrolysis reaction in locations other than at the surface of the substrate can be suppressed. Thus, a significant improvement in the rate of film formation and in the utilization factor of the vapor deposition reactant (metal compound) for film formation is achieved. Additionally, the uniformity of thickness and appearance of the oxide film is improved.

Claims 1, 5 and 8 stand finally rejected under 35 U.S.C. §103(a) as being unpatentable over U.S. Patent No. 5,919,726 to Hatano et al. (hereinafter “Hatano”) in view of U.S. Patent Application Publication No. 2002/0106321 to Tanaka et al. (hereinafter “Tanaka”)

The Office Action maintains the position that Hatano teaches a method for producing a photocatalyst material essentially as claimed, except for a three-second vapor mixing time. In response to Applicants’ arguments, the Office Action further states that col. 3, lines 26-31 of the reference teaches that a water vapor is sprayed concurrently with titanium tetrachloride vapor and, therefore, would mix such that “the mixed vapors are brought into contact with a substrate for vapor deposition”. The Examiner finally asserts that “...reaching the substrate with mixed vapors within 3 seconds of mixing is *reasonably expected* within the teaching of Hatano”. (Emphasis added.)

Applicants respectfully disagree. In Hatano, the water vapor sprayed during vapor deposition is supplied for recovery of the excess or unreacted TiCl_4 by dissolution to form an aqueous solution and *does not* participate in the vapor deposition reaction. In order to further distinguish the present invention from Hatano, claim 1 has been amended to recite that the hydrolysable metal compound vapor and the water vapor are reactant with one another.

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Furthermore, as argued in the previous response, Hatano fails to teach the claimed feature of pre-mixing a vapor of a hydrolysable metal compound and water vapor and bringing the mixed vapors into contact with the substrate within three seconds after mixing. Hatano fails to provide any teaching whatsoever that would lead one having ordinary skill in the art to “reasonably expect” that the vapors would come into contact within 3 seconds of mixing.

Also, as argued in the previous response, Hatano does not teach the vapor deposition defined in claim 1 in which a vapor of a hydrolysable metal compound and water vapor are brought into contact with a substrate to form a film of a metal oxide precursor on the surface of the substrate, and particularly in such a manner that the hydrolysable metal compound vapor and the water vapor are previously mixed before they are brought into contact with a substrate. Hatano actually teaches that water vapor, which is reactant with the titanium tetrachloride, is applied during a first step during silanol application and the titanium tetrachloride is applied in a second step without water vapor. In particular, Hatano teaches that during the coating method, the titanium tetrachloride is mixed with oxygen or dried air and reaction occurs on a coated surface.

In the process disclosed by Hatano, a silica gel-containing undercoating which contains silanol (-Si-OH) groups is formed in a first step. This undercoating can be formed by coating a substrate with a mixture of silica gel and water (column 2, lines 27-43). In the first step, a water vapor may be positively supplied to the atmosphere so as to form silanol groups sufficiently (column 3, lines 17-20).

In a second step, a titanium tetrachloride vapor is brought into contact with the undercoating having silanol groups, whereby it is vapor-deposited on the undercoating by a condensation reaction with the silanol groups on the undercoating as discussed in column 3, lines 21-22. The contact can be performed by bubbling liquid TiCl_4 with oxygen or dry air and atomizing or spraying the resulting TiCl_4 vapor onto the undercoating. Optionally, a water vapor may be sprayed to recover the excess TiCl_4 vapor as an aqueous solution after or during the supply of the TiCl_4 vapor (column 3, lines 21-58).

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Thus, in the process disclosed by Hatano, vapor deposition takes place by a condensation reaction between TiCl_4 vapor and silanol groups which have been previously formed on the substrate, and no water vapor is necessary for vapor deposition. Although Hatano teaches that a water vapor may be sprayed “during or after” vapor deposition, this water vapor is supplied for *recovery of the excess or unreacted TiCl_4 by dissolution to form an aqueous solution and does not participate in the vapor deposition reaction (condensation reaction)*. Such dissolution of TiCl_4 to form an aqueous solution interferes with a vapor deposition reaction. In a CVD technique, unreacted or excess reactant vapors should normally be discharged or expelled from the vapor deposition chamber as they are without condensation.

In any event, as stated above, claim 1 has been further amended to specifically recite the feature that the hydrolysable metal compound vapor and the water vapor are **reactant with one another**.

Accordingly, Hatano fails to teach at least the following features of claim 1 of a hydrolysable metal compound vapor and a water vapor which are reactant with one another are previously mixed and the mixed vapors are brought into contact with a substrate for vapor deposition; and the contact of the mixed vapors with substrate occurs within three seconds after mixing.

With respect to Tanaka, the Office Action continues to rely on Tanaka, directed to the production of ultra-fine particulate titanium oxide particles obtained from titanium tetrachloride by a vapor phase process, for teaching a three-second reaction/mixing time for the water vapor and titanium tetrachloride vapor. In response to Applicants’ arguments, the Office Action states that Tanaka is only relied upon as teaching a reaction time of three seconds or less and the fact that Tanaka does not teach depositing a film is “not relevant” as Hatano was utilized for this limitation.

Applicants respectfully disagree.

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If the Office Action relies on Tanaka as teaching a particularly claimed reaction time of two materials prior to application to a substrate, then the relied upon reference must be directed to the same materials being applied to the substrate. In contrast with the present invention, Tanaka relates to the production of **particulates**, not a deposited film. In particular, Tanaka teaches reacting titanium tetrachloride with an oxidizing gas, which may be steam, at a high temperature for a residence time of three seconds or less to obtain **particulates**. Tanaka is silent with respect to the step of coating a substrate with the mixed metal oxide film, let alone specifically teaching that the mixed vapors are brought into contact with the substrates *within three seconds after mixing*.

As discussed on page 4 of the present application, the inventors of the present invention have found that by bringing TiCl_4 vapor into contact with a substrate within three seconds, and preferably within one second, from its mixing with water vapor, the occurrence of a hydrolysis reaction of TiCl_4 in a location other than at the surface of the substrate can be suppressed. Thus, a significant improvement in the rate of film formation and in the utilization factor of TiCl_4 for film formation is achieved, and it is made possible to form a titanium oxide film having uniform thickness and good appearance. This passage in the specification continues on to state that it is thought that this is achieved because the vapors are mixed only on a macroscopic scale, but not completely mixed on a microscopic scale, which suppresses the reaction between TiCl_4 and H_2O and that TiCl_4 vapor contacting the substrate is increased, which contributes to an improved rate of film formation. None of the art of record recognizes the need for mixing the hydrolysable metal vapor with water vapor and bringing the mixture into contact with a substrate within three seconds from mixing for improved film formation.

As previously argued, Tanaka teaches the production of particulate titanium oxide by reacting a titanium tetrachloride-containing gas and an oxidizing gas, both preheated to 900°C or higher, with a residence time in the temperature range above 700°C which is controlled to be three seconds or less. The oxidizing gas may be steam (water vapor).

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The product produced by Tanaka is particulate titanium oxide, rather than a titanium oxide film, as produced by the present invention. In the production of a particulate titanium oxide, the reaction occurs in the gaseous (vapor) phase to form solid fine particles in the absence of a substrate, and all the gases which have reacted in this manner constitute the product.

The restriction of three seconds or less in the residence time above 700°C is for the purpose of controlling the growth of particles in order to obtain an ultra-fine particulate product ([0043]).

However, in vapor deposition or CVD technique, to form a titanium oxide film, such a reaction occurring in the vapor phase to form a fine powder is undesirable, since it leads to a wasteful consumption of TiCl_4 vapor (a decrease in film formation rate and in % utilization of TiCl_4 for film formation) and the resulting fine powder may adversely affect the film thickness uniformity and appearance of the titanium oxide film which is formed, as discussed on page 2, lines 10-31 of the present specification.

In the present invention, in order to control such undesirable or premature reaction between TiCl_4 vapor and water vapor occurring in the vapor phase to form a particulate solid, the two vapors are mixed within three seconds before contacting the substrate.

Thus, the reaction which is performed by Tanaka to form a particulate titanium oxide is undesirable in the film formation by a CVD technique according to the present invention, and the problems to be solved by the present invention discussed on page 2 of the specification do not cause any problem in the production of a particulate titanium oxide as disclosed by Tanaka.

Of further note, the temperature of the gases used in Tanaka (at least 900°C) is extremely high. One skilled in the art of CVD would not use vapors at such high temperature in the vapor deposition step of claim 1 to form a precursor film on a substrate by a CVD technique.

Additionally, there is no rationale to combine the teachings of Hatano and Tanaka. One having ordinary skill in the art would have no reason to combine the teachings of Tanaka (residence time) for producing a particulate titanium oxide by a reaction in the gaseous phase with the teachings of Hatano for producing a film of titanium oxide by a kind of CVD technique, since the formation of particulate titanium oxide (per Tanaka) by a reaction in the

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gaseous phase is undesired in producing a film.

Furthermore, even if the teachings of the two references could be combined, the combination does not lead to the subject matter of claim 1. First, Hatano does not teach previous mixture of a titanium tetrachloride vapor and a water vapor as reactants to form a film of a titanium oxide precursor as in claim 1. Second, Tanaka does not teach that mixed vapors of titanium tetrachloride and water vapor (steam) are brought into contact with a substrate within three seconds after mixing. Tanaka merely teaches that the residence time of mixed gases in the temperature range above 700°C is limited to three seconds or less in order to suppress the growth of the formed particles. **These teachings have no or little relevance to vapor deposition for film formation.**

In any event, claim 1 has been further amended to incorporate the features of dependent claim 2.

Claim 2 stands finally rejected under 35 U.S.C. §103(a) as being unpatentable over Hatano and Tanaka and further in view of U.S. Patent No. 4,293,326 to Terneu et al. (hereinafter "Terneu")

In the rejection of claim 2, Terneu is relied upon to teach a coating process whereby a vapor of a hydrolysable metal compound and water vapor are brought into contact in the vicinity of a traveling substrate via gas streams. In order to achieve the previous mixing of the vapors within three seconds before contacting the substrate, two jetted vapor streams meet each other before they reach the substrate. As shown in Fig. 1 of the present application, two lines are formed by extensions of the nozzles for making the vapor streams intersect with each other before reaching the substrate. However, the apparatus shown in the sole figure of Ternue is different in this respect. The two nozzles (11, 12) of Terneu are both directed on the glass ribbon (1) and do not meet each other before they reach the glass ribbon (1). Thus, the feature of previously rejected claim 2 of two vapor streams meeting each other before reaching the substrate is not considered by Terneu. There is no reason to modify Terneu to change the directions in which the nozzles (11, 12) are directed, since pre-mixing of the vapors is not relevant to the Terneu process.

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In response to this argument, the Office Action asserts that metal vapor and water vapor are supplied in separate streams and they “come into contact in the vicinity of the substrate” and thus Applicants’ argument is “without merit”. The Examiner’s attention is directed to column 5, lines 40-52 of Terneu, which specifically states that the discharge ends of the conduits open into a shallow flow passage 13 defined by a shroud 14 and by the **top face of the glass ribbon** and that the inclined discharge end portions of the conduits define passageways from which the gas streams issue in the **form of layers** extending over substantially the width of the flow passage. In other words, the conduits apply the gas streams *in layers* onto the glass ribbon and do not meet and mix prior to contact with the glass ribbon.

In view of the amendment to claim 1, which requires the features of previously filed dependent claim 2, that the vapor deposition step is carried out by injection of jetted streams of the hydrolysable metal compound vapor and water vapor toward the substrate which is continuously moving, and the mixing is performed by injecting the hydrolysable metal compound vapor and the water vapor in such a manner that the resulting two jetted vapor streams meet each other before they reach the substrate and in view of the reasons set forth above, it is respectfully requested that the rejection of amended claim 1 under 35 U.S.C. §103(a) be withdrawn as the combination of Hatano, Tanaka and Terneu fails to render this claim obvious.

For the reasons set forth above, a *prima facie* case of obviousness against independent claim 1 has not been established.

Claims 3-4 are finally rejected under 35 U.S.C. §103(a) as being unpatentable over Hatano, Tanaka, Terneu and further in view of WO 89/00549 to Lindner (hereinafter “Lindner”).

Lindner is relied upon as teaching the use of slit nozzles arranged in opposing relation to and toward each other at an angle for use in a coating process. Claims 3 and 4 depend directly from amended claim 1 and add further limitations to claim 1 and are believed to be allowable for at least the reasons discussed hereinabove. Lindner fails to overcome the deficiencies of the Hatano, Tanaka and Terneu combination in the rejection of amended claim 1.

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Furthermore, Lindner does not teach that two vapors are previously mixed before contacting a substrate or that two jetted vapor streams meet each other before reaching the substrate as defined in claim 1. This is apparent from Figs. 9 and 17 of Lindner. The two supply nozzles (10a, 10b) are angled with respect to each other, but they do not deliver vapor streams that meet each other before reaching the substrate (12).

For the reasons set forth above, it is respectfully requested that the rejection of claims 3-4 under 35 U.S.C. §103(a) be withdrawn as the combination of Hatano, Tanaka, Terneu and Lindner fails to render these claims obvious.

Claims 6 and 7 are finally rejected under 35 U.S.C. §103(a) as being unpatentable over Hatano and Tanaka and further in view of U.S. Patent No. 4,261,722 to Novak et al. (hereinafter "Novak").

Novak is relied upon as teaching a coating process wherein the temperature of the substrate to be coated may be in the range of 150-700°C. Claims 6 and 7 depend directly from claim 5 and add further limitations to this claim and independent claim 1 and are believed to be allowable for at least the reasons discussed hereinabove in connection with independent claim 1 and dependent claim 5. Novak fails to overcome the deficiencies of the Hatano and Tanaka combination in the rejection of claims 1 and 5.

For the reasons set forth above, it is respectfully requested that the rejection of claims 6-7 under 35 U.S.C. §103(a) be withdrawn as the combination of Hatano, Tanaka and Novak fail to render these claims obvious.

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CONCLUSION

In view of the arguments set forth above and the amendments to the claims, it is respectfully requested that this Amendment be entered and the application passed to issue.

Respectfully submitted,

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